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CROSS-LINKED PRODUCTS WITH NO BLOOMING AND REDUCED
FOGGING*5
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The present invention relates to compositions comprising peroxide and a specific copolymer, compositions comprising said copolymer and carrier materials, a cross-linking process of polymers/elastomers involving the use of peroxides and said copolymer, and the products obtained by this process. The resulting (cross-linked) products show essentially no blooming, reduced emission of volatile compounds, and less fogging. Preferably, the cross-linking is predominantly of a polyolefin or elastomer through reactions initiated by radicals obtained by decomposition of the peroxide. The resulting cross-linked products can be in the form of finished articles or intermediates.

Typically, products that are obtained through a peroxide initiated cross-linking process suffer from blooming of the decomposition products of said peroxide. Furthermore, said decomposition products can be quite volatile in nature and hence be emitted from the cross-linked article/intermediate, which can lead to fogging, e.g., on automobile windows. Since such blooming/emission is undesired, several solutions have been put forward to reduce these phenomena. US patent No. 3,420,891, for instance, proposes to prevent blooming by using certain tris-peroxides. Belgium patent No. 883.693, on the other hand, proposes to use phthalic anhydride in an elastomer composition. According to a non-binding theory as described in this patent, the phthalic anhydride reacts with an OH group-bearing decomposition product of the peroxide. While the OH group-bearing decomposition product would lead to blooming, the reaction product with the phthalic anhydride does not. The phthalic anhydride was used in a quantity of at least 2 moles per mole of (difunctional) peroxide used.

The use of phthalic anhydride, however, has its drawbacks. More particularly, the quantity of OH group-bearing peroxide decomposition products formed will depend on the reaction conditions when the peroxide
5 decomposes. If the reaction conditions are such that the radicals undergo β -scission, for example, a significantly reduced quantity of OH group-bearing products is formed (i.e. because ketones are formed). Similarly, if ethylenically unsaturated species are present when the peroxide decomposes, the radicals may react with the unsaturated moiety, resulting
10 in fewer OH group-bearing decomposition products. Also, "in-cage" termination reactions might occur, wherein two radical fragments of the peroxide, after one or more rearrangements, terminate with one another. Hence, the quantity of phthalic anhydride needed to react with the OH group-bearing peroxide decomposition products that are formed is
15 dependent on many factors that, more often than not, cannot be controlled beforehand. Consequently, there will hardly ever be stoichiometric quantities of phthalic anhydride and OH group-bearing peroxide decomposition products.

20 Unfortunately, it is not only the peroxide decomposition products that may bloom or be emitted from the cross-linked article/intermediate. Also phthalic anhydride was observed to cause blooming. As a result, products obtained by a process comprising a peroxide-initiated cross-linking step of a polymer/elastomer composition comprising phthalic anhydride typically will
25 either suffer from blooming/emission of (OH group-bearing) peroxide decomposition products, or from blooming of excessive phthalic anhydride.

Furthermore, the industry is looking for a solution to the blooming problem that has a good price/performance ratio and, preferably, allows the use of

conventional peroxides, such as dicumyl peroxide, bis(tert-butylperoxyisopropyl)benzene, 2,5-bis(tert-butylperoxy)-2,5-dimethylhexane, 2,5-bis(tert-butylperoxy)-2,5-dimethyl-3-hexyne and/or tert-butyl cumyl peroxide, since these peroxides have proven themselves over the years. More preferably, the solution to the blooming problem should not interfere with the cross-linking process so that recipes do not have to be changed other than in adding a certain ingredient.

There has been an ongoing search for peroxide-containing compositions that will satisfy those needs. Surprisingly, we have found that the combination of peroxides and specific olefinic copolymers according to the invention is particularly suitable, and that the use of such a combination leads to a cost-efficient cross-linking process without significant recipe changes being necessary. The resulting products show essentially no blooming, and the emission of volatile matter from the product as well as fogging caused by the product was reduced in comparison with products wherein the peroxide was used without said polymer/elastomer being present.

The invention is characterized in that peroxide is used in combination with one or more copolymers comprising olefin-derived moieties and one or more anhydride and/or acid groups, the copolymer being used in a quantity of from 0.1 to 500 per cent by weight of the weight of the peroxide.

It is noted that Japanese patent application No. 08157646 discloses the use of such copolymers in elastomer compositions that contain brass and/or brass-plated iron in order to improve the adhesion of the elastomer, after peroxide-initiated vulcanization, to said brass. Such compositions are not the subject of the present invention. Furthermore, this Japanese patent

application is silent with respect to blooming, the emission of volatile products and/or fogging.

The copolymers containing olefin-derived moieties and anhydride and/or acid groups can be suitably obtained through (random) copolymerization or grafting processes. EP-A-0 429 123, for instance, discloses a suitable process wherein olefins (1), or mixtures of olefins, are reacted with α,β -unsaturated dicarboxylic compounds (2), in the presence of a polymerization inhibitor, and preferably in the substantial absence of the well-known polymerization initiators, at temperatures from about 180°C to about 350°C and at subatmospheric, atmospheric or superatmospheric pressures, but preferably at atmospheric pressure. The reaction is preferably carried out in the absence of oxygen, i.e. under an inert (nitrogen) atmosphere. Reaction times can range from about 0.5 to about 15 hours. The inhibitor utilized is said to be used preferably in a quantity of from about 0.01% to about 3% by weight, based on the combined weight of components (1) and (2). Components (1) and (2) may be reacted in a single dose or may be dosed continuously or periodically throughout the reaction cycle. The copolymers so obtained may additionally be hydrogenated, for example by heating at 130 – 200°C in the presence of a catalyst, e.g., nickel or palladium. Such hydrogenated compositions typically have improved thermo-oxidative stability.

Typical examples of preferred copolymers as used according to the present invention are Dapral® PX 200 and Ricobond® as supplied by Akzo Nobel and Ricon Resins Inc., respectively.

Olefins suitable for use in the copolymer of the invention include propylene, butadiene, isobutylene, and C₆ to C₂₄ α -olefins. Preferred olefins for use in

these copolymers have 12 to 18 carbon atoms. More preferably, at least one C₁₂ to C₁₈ α -olefin, such as dodecene-1, tridecene-1, tetradecene-1, pentadecene-1, hexadecene-1, heptadecene-1, and octadecene-1, is used.

- 5 Preferred unsaturated anhydride and/or acid moiety-containing compounds used to introduce anhydride and/or acid groups into the copolymer include maleic anhydride; (di)alkyl/aryl-maleic anhydride, such as n-dodecyl maleic anhydride, citraconic anhydride, and maleic phenyl anhydride; (substituted) norbornene-2,3-dicarboxylic anhydride; (meth)acrylic acid; maleic acid;
10 fumaric acid; itaconic acid; citraconic acid; and other anhydrides and/or acids with an unsaturated group that can be linked with an olefin.

Preferably, the copolymer contains anhydride and/or acid groups derivable from maleic anhydride or methacrylic acid. The copolymers might also be
15 adducts of alkadiene polymers and cyclic or acyclic anhydrides of mono- or dicarboxylic acids containing 4 to 18 carbon atoms as disclosed in US patent No. 4,423,177.

Although it is not critical, the copolymers preferably have a composition as
20 obtainable by reacting one or more olefins and one or more anhydride/acid group-containing monomers, with the anhydride/acid group-containing monomers being present in a quantity of 5-75 per cent by weight (%w/w). More preferably, the anhydride/acid group-containing monomers are present in a quantity of 10-50 %w/w.

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The copolymers were found not to bloom from products comprising them, possibly due to their molecular weight. To avoid blooming, the molecular weight of the copolymer, most likely, will have to be over 500, preferably over 800, and more preferably over 1100 Dalton. However, in order to

facilitate processing of compositions comprising the copolymer, and to allow for sufficient mobility of the copolymer in the final matrix, the molecular weight of the copolymer must be below 50,000, preferably below 25,000, and more preferably below 15,000. Typically, an alternating
5 copolymer with 5 to 50 repeating units can suitably be used.

The quantity of said copolymer that is to be used in the peroxide-containing elastomer/polyolefin compositions can vary over a wide range and can be optimized on the particular composition and the conditions during peroxide
10 decomposition. However, since the copolymer does not bloom at all, it can be used in a more than stoichiometric quantity, based on the quantity of OH group-bearing decomposition products that can be formed by the peroxide (by radical abstraction). Because it is estimated that as little as 50% of the radicals from the decomposing peroxide might actually lead to
15 the formation of OH group-bearing decomposition products, the minimum quantity of copolymer to be used in the peroxide-containing elastomer/polyolefin compositions of the invention is such that the total number of anhydride/acid groups of the copolymer equals 50% of the maximum quantity of OH groups that can be formed from the decomposing
20 peroxide. Typically this means that at least 0.1 %w/w of copolymer is present, based on the weight of the peroxide. The maximum quantity of copolymer to be used depends on how much of it is tolerated in the product and on economic considerations. Typically, less than 500%w/w is used, based on the weight of the peroxide, since otherwise the mechanical
25 properties of the final, cross-linked product may deteriorate. Preferably, the copolymer is present in a quantity of 1-400 %w/w, more preferably in a quantity of 10-300 %w/w, based on the weight of the peroxide. Alternatively, one can specify the quantity of copolymer to be used on the weight percentage that ends up in the cross-linked product. In that case,

from 0.05 to 10 %w/w of copolymer, preferably 0.5 to 7.5 %w/w, is present in the product.

The polymer/elastomer can be cross-linked in a conventional way using a peroxide in combination with the copolymer. The copolymer and the peroxide may be blended/mixed together or separately with the polymer/elastomer at any time using conventional equipment, such as a (twin-screw) extruder. Preferably, they are blended/mixed into the polymer/elastomer together and jointly with optional further ingredients, so that just one mixing step is required. However, differences in viscosity of the compounds to be blended/mixed may dictate otherwise. Large viscosity differences, for example, may require that one of the components be introduced in a stepwise fashion to ensure efficient mixing/blending.

It is noted that the term "polymer/elastomer" as used throughout this document is meant to denominate all thermoplastic and/or rubbery polymers. Preferably, the polymer/elastomer is a (co)polymer of ethylene, propylene and/or a synthetic or natural rubber. Examples include, but are not limited to, polypropylene (PP) and/or one of the more preferred polymers/elastomers selected from the group consisting of polyethylene (LDPE, LLDPE, HDPE); chlorinated polyethylene; ethylene- α -olefin copolymers, such as ethylene/propylene copolymers (EPM and EPDM), ethylene octene copolymer (EOP); ethylene-vinyl acetate copolymer (EVA); polyacrylic acid ester; natural rubber (NR); polybutadiene rubber (BR); polybutylene; polyisobutylene; styrene butadiene rubber (SBR); acrylonitrile-butadiene-styrene terpolymer (ABS); (acrylo)nitrile butadiene rubber (NBR); hydrogenated (acrylo)nitrile butadiene rubber (HNBR); polyisoprene; polychloroprene rubber (CR); silicone rubber (VMQ, MQ); urethane rubber; fluoro elastomers (FKM), including fluorinated rubber; and

thermoplastic vulcanizates (TPV). More preferred types of polymer/elastomer include polyethylene, ethylene-vinyl acetate copolymer, butadiene-acrylonitrile copolymer, EPM; EPDM, such as Vistalon®606 and Vistalon®7500 ex Exxon and Buna®EP 6550 and Buna®EP G8450 ex
5 Bayer; EOP, such as Engage®8150 and Engage®8180 ex DuPont-Dow-Elastomers; VMQ, such as Silopren®HV ex Bayer; and FKM, such as Viton® products ex DuPont-Dow-Elastomers, all of which are low-priced, widely available, and have excellent physical properties that allow wide-ranging use. Most preferred are compositions based on synthetic rubbers,
10 since here the blooming phenomenon is most pronounced, such as EPM, EPDM, and EOP.

The term "cross-linking" is used in the usual way, meaning that a three-dimensional network of polymer chains is formed as is detailed in, for
15 instance, chapter 1.3 of W. Hofmann's *Rubber technology handbook* (Carl Hanser Verlag, 1989). The term does not comprise the very different grafting process such as described in FR-A-2 559 774, wherein a monomer unit is grafted onto a polymer to increase its adhesion to polar polymers and/or metals.

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It is furthermore to be understood that the term "blooming" is used as it is conventionally in the art. Blooming typically is the result of products migrating to the surface of a polymeric/elastomeric article. Whether a product is subject to blooming or not depends on the solubility of the
25 compound and the rate of diffusion. As is conventional, products are considered not to bloom when no changes on the surface are observed (visual inspection) upon storage for a prolonged time at temperatures above the temperatures that may be encountered during projected use of the products. To determine whether the cross-linked polymeric/elastomeric

article emits volatile compounds over time, conveniently use is made of a fogging test.

The peroxide as used in the compositions according to the invention can be any conventional peroxide. Preferably, the peroxides are chosen from products conventionally used in cross-linking and degradation reactions, such as dialkyl peroxides, perketals, peroxyarbonates, and the like. More preferably, the peroxide or mixture of peroxides that is used comprises a peroxide selected from the group consisting of bis(tert-butylperoxyisopropyl)benzene, "liquified" versions of this peroxide, meaning that one or more tert-butyl groups are replaced by other (more) bulky groups or that may contain substituents on the benzene ring, dicumyl peroxide, 2,5-bis(tert-butylperoxy)-2,5-dimethylhexane, 2,5-bis(tert-butylperoxy)-2,5-dimethyl-3-hexyne, 1,3,5-tris(tert-butylperoxyisopropyl)benzene, tert-butyl cumyl peroxide, di-t-butyl peroxide, 1,1-bis(tert-butylperoxy)-3,3,5-trimethylcyclohexane, 1,1-bis(tert-butylperoxy)cyclohexane, n-butyl-4,4-bis(tert-butylperoxy)valerate, ethyl-3,3-bis(tert-butylperoxy)butyrate, tert-butylperoxy 2-ethylhexyl carbonate, tert-butylperoxy isopropyl carbonate, tert-butyl peroxybenzoate, and less preferred peroxydicarbonates, such as bis(4-tert-butylcyclohexyl) peroxydicarbonate, and bis(2-ethylhexyl) peroxydicarbonate. The most preferred peroxides for use in the process according to the invention are the conventional ones, bis(tert-butylperoxyisopropyl)benzene in particular. In the degradation and cross-linking processes according to the invention, the amount of peroxide used preferably ranges from 1 to 5 per cent by weight, based on the weight of the polymer/elastomer to be degraded or cross-linked. More preferably, the peroxide is used in an amount of 1.5 – 4.0 per cent by weight, based on the weight of the polymer/elastomer.

In a further embodiment, the invention relates to compositions comprising peroxide and a copolymer with olefin-derived moieties and anhydride and/or acid groups, which compositions are pre-eminently suited for use in the cross-linking process. These compositions, if so desired, may contain
5 further conventional additives, such as plasticizers (e.g. paraffinic oils and/or esters), antioxidants, scorch retarders, and the like, and conventional carrier materials, such as silica and/or chalk. The conventional additives and carrier materials are presented in more detail below. Typically, such compositions will contain 5-60% by weight, based on
10 the total weight of the composition, of peroxide, 0.1-500% by weight, based on the weight of the peroxide(s), of the copolymer, optional further additives, and 0-50% by weight of carrier material, up to a total of 100%. Preferred compositions comprise one or more peroxides, copolymer with olefin-derived moieties and anhydride and/or acid groups, one or more
15 fillers selected from the group consisting of silica, clay, chalk, kaolin, and carbon black, and optional further conventional additives.

In a further embodiment, the invention relates to compositions comprising one or more of the conventional additives and/or conventional carrier
20 materials, as mentioned below, and copolymer with olefin-derived moieties and anhydride and/or acid groups. Typically, such compositions will contain 1-99.9% by weight, based on the total weight of the composition, of additives and/or carrier material and 99-0.1% by weight, based on the weight of the composition, of the copolymer, up to a total of 100%. More
25 preferably, the compositions will contain 10-99% of additives and/or carrier material and 90-1% of the copolymer. Even more preferred are compositions containing 25-90% of additives and/or carrier material and 75-10% of the copolymer. Preferred compositions comprise copolymer with olefin-derived moieties and anhydride and/or acid groups, one or more

fillers selected from the group consisting of silica, clay, chalk, kaolin, and carbon black, and optional further conventional additives mentioned above.

The products obtainable by this process, which show essentially no blooming and reduced emission of volatiles, are typically but not limited to: building profiles; automotive profiles, such as sponge door seals and weatherstrips; mechanical goods, such as hose, conveyor belt cover materials, and seals; and the like. Again, the products may be in the final form and shape, or be present as an intermediate that needs further processing. Such an intermediate may be a granulate of (partially) cross-linked polymer/elastomer.

In the process of the present invention, conventional adjuvants (additives) are optionally used. Examples of such additives include co-agents; monomers; chain transfer agents; initiators, such as NO compounds; fillers; stabilizers, such as inhibitors of oxidative, thermal, or ultraviolet degradation, also known as antioxidants, antidegradants and/or scorch retarders; lubricants; extender oils; pH controlling substances, such as magnesium oxide and calcium carbonate; release agents; vulcanizing agents, such as sulfur; colourants; plasticizers; diluents; accelerators; and the like. Examples of reinforcing or non-reinforcing fillers are: silica, clay, chalk, carbon black, and fibrous materials, such as glass fibres, and the like.

Experimental

Materials used:

Perkadox® 14 R – Di(tert-butylperoxyisopropyl)benzene (ex Akzo Nobel)

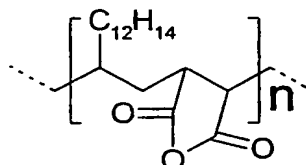
Perkadox® 14 40 B pd ex Akzo Nobel)

Keltan® 520 - EPDM (ex DSM)

Carbon black SRF N-772 (ex Cabot)

Sunpar® 150— paraffin oil (ex Sunoco)

Dapral® PX 200 – α -olefin maleic anhydride copolymer (ex Akzo Nobel) of



formula

wherein n is about 30.

5 Phthalic anhydride (ex Baker)

Cis-hexahydrophthalic anhydride (ex Acros Organics)

Succinic anhydride (ex Fluka Chemie)

Methylnorbornene-2,3 dicarboxylic anhydride (ex Acros Organics)

1,2,4,5 benzene tetracarboxylic anhydride (ex Acros Organics)

10 1,3,5-tris-(t-butyl-peroxyisopropyl)benzene (ex Akzo Nobel)

Abbreviations used:

MAH = maleic anhydride

MW = molecular weight (of anhydride)

15 Px-14R = Perkadox® 14 R, 50% by weight of peroxide.

Px-14 = Perkadox® 14 40 B pd, 40% by weight of peroxide

PX-200 = Dapral® PX 200

Procedures:

20 Unless stated otherwise, a standard 100 / 50 / 10 compound (100 parts by weight rubber, 50 parts by weight carbon black, and 10 parts by weight paraffin oil) was used as the polymer/elastomer for testing purposes. Compounding, in a 5-litre internal mixer ex Werner & Pfleiderer, of 1.6 kg Keltan® 520, 0.8 kg carbon black SRF N-772, and 0.16 kg Sunpar® 150,
25 produced this standard compound.

Further ingredients, i.e. peroxide and reactive additive, were used in said standard compound for evaluation. The ingredients were incorporated into the compound on a two-roll mill (Schwabenthal 150x350mm) at a starting temperature of about 50°C. The cross-linking of the resulting 2 mm rubber
5 sheet was done by compression moulding at 175°C for 15 minutes, in which step virtually all peroxide was decomposed.

The extractable matter in the rubber, after cross-linking, was determined by extraction with dichloromethane and subsequent GC-analysis. The quantity
10 of extractable matter is an indication of the likelihood that the cross-linked product will show blooming or emit volatile matter.

Some products were evaluated using dynamic head space GC analysis to determine the quantity of volatile matter in the product. To this end, a
15 sample of 30-50 mg of the product was heated in a glass tube under a stream of inert gas. The desorbed volatile compounds were trapped in a liquid nitrogen cooled trap placed downstream of the sample. After the heat cycle "desorption time" the cold trap was heated rapidly and the condensed volatile compounds evaporated. The compounds were transferred directly
20 to a capillary GC column and separated by means of normal GC analysis. The heating temperature/desorption time was either 90°C/30 minutes (cycle A) or 120°C/60 minutes (cycle B).

The total GC peaks surface area was tallied up and given as toluene equivalents (ppm toluene), being the quantity of toluene that would give the
25 same peak area when analysed under the same conditions.

In some other cases, the effect of volatile matter in cross-linked materials was analyzed by determining fogging in accordance with method DIN 75201 (gravimetrically).

The blooming of peroxide decomposition products and other ingredients on the surface of rubber sheets was determined visually. The sheets were stored in sealed glass bottles at room temperature. After a defined time
5 period (1 week / 1 month) the rubber sheets were inspected visually. If the surface of the rubber sheets remained glossy, they were said not to show blooming. When the surface was not glossy or was covered with crystals, the sheets were said to show blooming.

10 Example 1 and Comparative Examples A-F

In all examples 6.6 g of Px-14 R (9.75 mmol peroxide) were used per 160 g of standard compound and a further additive/copolymer was used as given in Table 1. The quantity added was again expressed in grams per 160 g of standard compound. "MAH (meq)" stands for the quantity of reactive
15 anhydride/acid groups, expressed in milli-equivalents, as introduced by the additive(copolymer).

Table 1

Ex.	Additive (Copolymer)	MW (Dalton)	Added (g)	MAH (meq)	Blooming of peroxide / additive	
					1 week	1 month
A	None	-	-	-	Yes	Yes
B	succinic anhydride	100	2	20	Yes	Yes
C	phthalic anhydride	148.1	2	13.5	Yes	Yes
D	1,2 4,5 benzene-tetra carbacid	254	2	15.7	Yes	Yes
E	cis-hexahydro phthalic anhydride	154	1.2	7.8	No	Yes
F	Methylnorbornene- 2,3 dicarboxylic anhydride	178	1.4	7.8	No	Yes
1	PX-200 1/1	(308) _n	3	9.75	No	No

- From the product of Example 1, in total about 1,000 ppm of $\text{CH}_3\text{C}(\text{O})-\text{C}_6\text{H}_4-\text{C}(\text{CH}_3)_2-\text{OH}$ and $\text{HO}-\text{C}(\text{CH}_3)_2-\text{C}_6\text{H}_4-\text{C}(\text{CH}_3)_2-\text{OH}$ could be extracted, whereas Comparative Examples A, E, and F showed extractable levels of these compounds of 3,500-5,500 ppm. Comparative Examples B-D showed low levels of extractables, due to the high amount of anhydride/acid used. However, in these tests the excess anhydride/acid bloomed from the product.
- Head space GC analysis using cycles A and B showed the product of Example 1 to contain about 500 and 1,700 ppm toluene, respectively, while the product of Comparative Example A contained 1,100 and 3,500 ppm toluene, respectively. Each number is the average result of two samples.

Examples 2-5 and Comparative Examples G and H

The previous examples were repeated, except that 4.2 g of Px-14 (4.96 mmol) were used per 160 g of standard compound. The total quantity of

toluene, respectively. Each number is the average result of two samples.

Examples 2-5 and Comparative Examples G and H

The previous examples were repeated, except that 4.2 g of Px-14 (4.96 mmol) were used per 160 g of standard compound. The total quantity of extractable $\text{HO-C(CH}_3)_2\text{-C}_6\text{H}_4\text{-C(CH}_3)_2\text{-OH}$ was also determined. The results are given in Table 2.

Table 2

Ex.	Copolymer	Added	MAH (meq)	Blooming of peroxide / additive		Extraction (ppm)
				1 week	1 month	
G	None	-	-	Yes	Yes	1210
2	PX-200 1/1	1	3.25	No	Yes	375
3	PX-200 1/1	2	6.5	No	Yes	105
H	None	-	-	Yes	Yes	1740
4	PX-200 1/1	3	9.75	No	No	70
5	PX-200 1/1	4	13.0	No	No	40

10

Clearly, the combination of peroxide and copolymer leads to a reduction of volatile/extractable material in the cross-linked product, and non-blooming products can be obtained. The quantity of copolymer needed to obtain non-blooming formulations will be dependent on the actual reaction conditions, but can be evaluated simply by the above-mentioned method.

15

Head space GC analysis using cycles A and B showed the product of Example 3 to contain 550 and 1,850 ppm toluene, respectively, whereas the product of Comparative Example G contained 700 and 2,000 ppm toluene, respectively. Similarly, the product of Example 4 contained 350

Again, each number is the average result of two samples.

Fogging tests of the cross-linked products of Examples G, 3, and 4, and H and 4 (in two series of tests) showed the following:

Example	Fogging condense (mg)	Fogging reduction (%)
G	38	n.r.
3	25	34
4	22	42
H	42	n.r.
4	23	45

- 5 Clearly a substantial reduction of fogging is reached with the process according to the invention.

Example 6 and Comparative Example I

10 The previous examples were repeated, except that 2.4 g of 1,3,5-tris-(t-butyl-peroxyisopropyl)benzene (5 mmol) were used per 160 g of standard compound. In Comparative Example I no copolymer was used, while Example 6 used 2 g of PX-200 1/1. After one month the product of Comparative Example I showed blooming, while the product of Example 6 did not.

- 15 When analysed for extractable/volatile matter, the product of Example 6 showed that a total of 2,180 ppm of $C_6H_3-(C(CH_3)_2-OH)_3$ and $CH_3C(O)-C_6H_3-(C(CH_3)_2-OH)_2$ could be extracted, while the product of Comparative Example I contained 4,500 ppm of such extractable compounds.

20 Head space GC analysis using cycles A and B showed the product of Example 6 to contain 150 and 850 ppm toluene, respectively, while the product of Comparative Example I contained 165 and 1,700 ppm toluene,

respectively. Each number is the average result of two samples.